Supplemental Material

Temperature- and depth-dependent valence band electronic structures of half-metallic Co_2MnSi studied by hard x-ray photoemission spectroscopy

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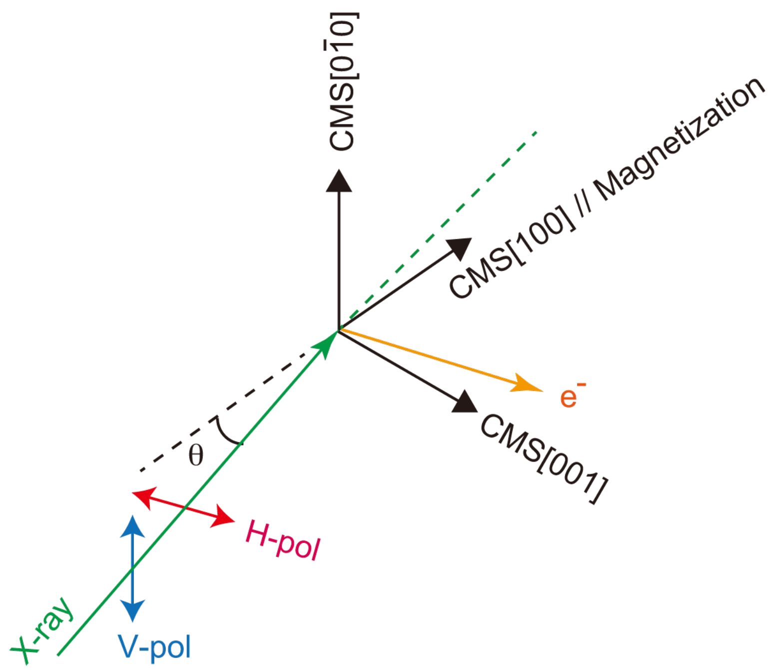

FIG. S1. Schematic diagram of the experimental geometry of HAXPES measurements. See also Ref. [25]. The incidence angle of x-ray relative to the CMS(001) plane is indicated by θ in the figure.

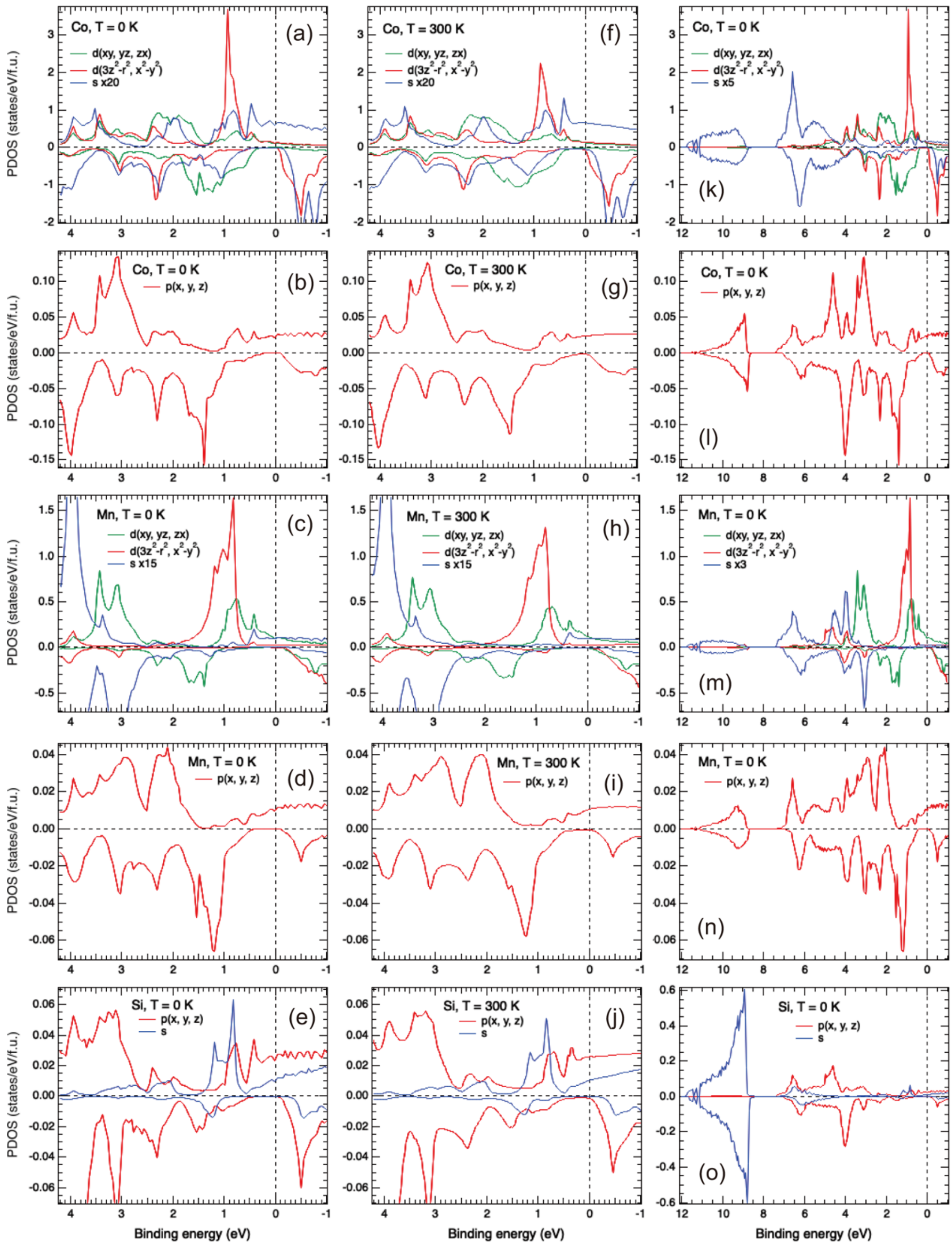

FIG. S2. Calculated spin-resolved partial densities of states (PDOSs) near the Fermi level of $L2_1$-ordered Co_2MnSi (CMS) at temperatures (*T*) of 0 K (a)-(e) and 300 K (f)-(j). (k)-(o) PDOSs in the entire valence band region for (a)-(e). The PDOS data are obtained from Ref. [9].

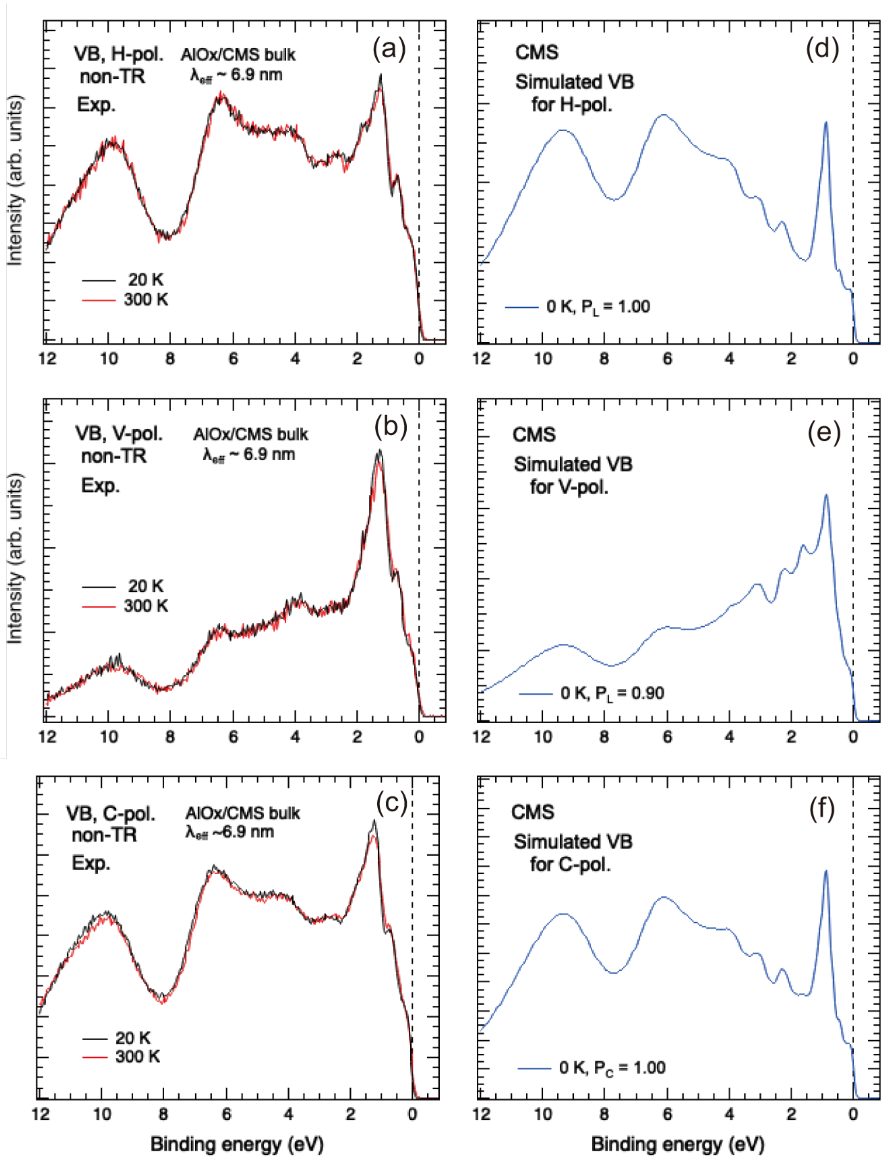

FIG. S3. (a)-(c) Experimental HAXPES spectra in the entire valence band (VB) region measured at T = 20 and 300 K for H-, V-, C-pol x-rays, respectively. An integrated-type background was subtracted in each experimental spectrum. (d)-(f) Simulated HAXPES spectra with using the PDOSs for H-, V-, and C-pol x-rays at T = 0 K. In the simulations, the cross-sections including the matrix element and photoelectron diffraction effects and the degree of x-ray polarization (P_L or P_C) for each polarization are taken into account. The experimental spectrum measured at T = 300 K for C-pol x-rays was taken from Ref. [20].

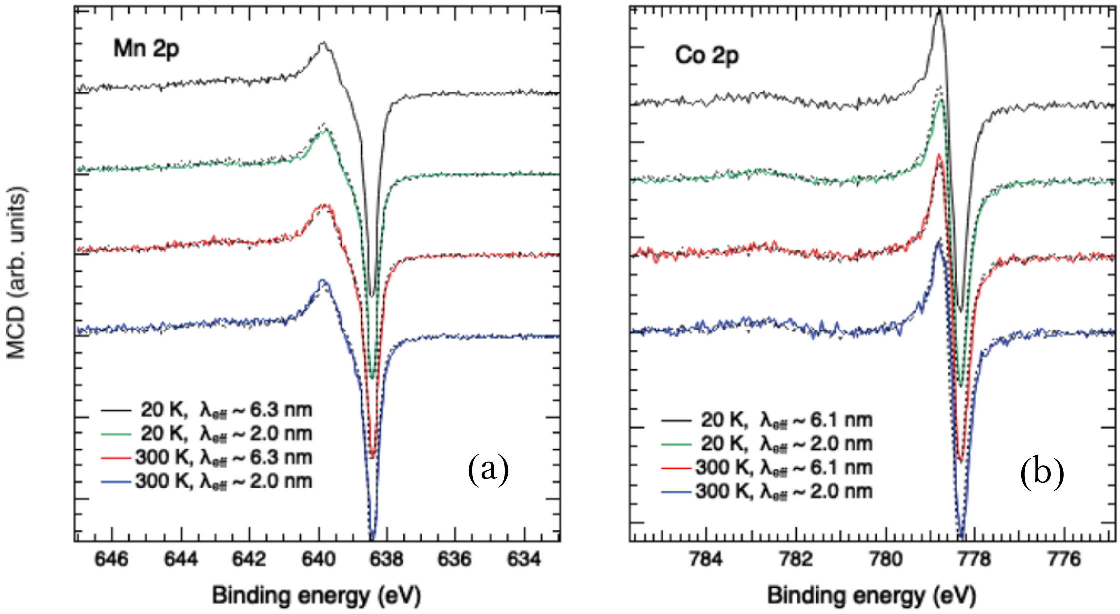

FIG. S4. (a) Normalized Mn $2p_{3/2}$ core-level MCD profiles in HAXPES at $T = 20$ and 300 K for bulk ($\lambda_{eff} \sim 6.3$ nm) and near-interface ($\lambda_{eff} \sim 2.0$ nm). (b) Normalized Co $2p_{3/2}$ core-level MCD profiles in HAXPES at $T = 20$ and 300 K for bulk ($\lambda_{eff} \sim 6.3$ nm) and near-interface ($\lambda_{eff} \sim 2.0$ nm). The MCD profiles at $T = 300$ K were obtained from Ref. [20].

Satellite structure in the Co 2*p* core-level HAXPES

In this section, we consider the origin of the satellite structure, which is indicated by the red arrows, in the Co 2*p* HAXPES spectra shown in Figs. 1(a) and 1(b). The satellite in the $2p_{3/2}$ region shows a clear hump structure, while that in the $2p_{1/2}$ region is broadened by a shorter lifetime of the $2p_{1/2}$ core-hole than the $2p_{3/2}$ one. This hump structure has been commonly observed in Co-based Heusler alloys [47-50], but the origin of the hump structure is unclarified yet. As a possible origin of the satellite, shake-up transition and/or energy loss process in the photoemission final states has been mentioned in Refs. [47-50]. A similar satellite structure can be seen in the 2*p* core-level PES for metallic Ni (so-called 6-eV satellite), and has been interpreted by the charge transfer satellite in the photoemission final states for Ni metal [28]. In the case of the charge transfer satellite, the Ni 2*p* core-level magnetic dichroism in PES shows the positive-to-negative sign change in both the main and satellite structure in the $2p_{3/2}$ region and shows the opposite sign change in the $2p_{1/2}$ region [29-32]. As shown in Fig. 1(e), the sign of Co 2*p* MCD in HAXPES is positive (negative) in the $2p_{3/2}$ ($2p_{1/2}$) region as indicated by the red arrows. No sign reversal in MCD in the $2p_{3/2}$ ($2p_{1/2}$) satellite region strongly suggests that the origin of the hump in the Co 2*p* HAXPES for CMS is not due to the charge transfer satellite. In the case of the energy loss process as an origin of the hump, the hump is created by the energy loss replica of the main peak, and the MCD for the hump would also show the replica of MCD for the main peak. The shake-up process might give a replica of the main peak as a hump structure as well as the energy loss process. Therefore, the origin of the hump might be due to neither the shake-up nor energy loss process, which is confirmed by no sign reversal in MCD for the hump as seen in Fig. 1(e). We thus suspect that a possible origin of the hump in the Co 2*p* HAXPES spectra in CMS is the multiplet structures in the photoemission final states, since the multipets in the photoemission final states cannot be ignored for a metal with relatively localized 3*d* orbital. As seen in Figs. 1(c) and 1(d), the Mn 2*p* HAXPES spectra show the broad tails indicated by the blue arrows in the higher E_B side of the main peak in both the $2p_{3/2}$ and $2p_{1/2}$ regions. The sign of MCD in the broad tail is positive (negative) in the the $2p_{3/2}$ ($2p_{1/2}$) region as shown in Fig. 1(f). In addition, the Fe 2*p* HAXPES spectra for the Fe thin films [33] also show a similar broad tail and a positive (negative) sign of MCD in the higher E_B side of the $2p_{3/2}$ ($2p_{1/2}$) main peak. These facts suggest the presence of the mutliplet states in the higher E_B side of the main peak. For the Co 2*p* HAXPES, the hump is observed instead of the broad tails in the Mn and Fe 2*p* HAXPES probably due to the variations of multiplet states depending on the number of 3*d* electrons. To further discuss the origin of the hump in the Co 2*p* HAXPES, the theoretical approach, which correctly treats the band structure and the core-level photoemission final states for ferromagnetic metal systems, is required.